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REPUBLIC OF SOUTH AFRICA  
PATENTS ACT, 1978  
APPLICATION FOR A PATENT AND  
ACKNOWLEDGEMENT OF RECEIPT  
(Section 30(1) Regulation 22)

REPUBLIC OF SOUTH AFRICA  
FORM P.1 REVENUE  
(to be lodged in duplicate)

-910.98

R 266.00

THE GRANT OF A PATENT IS HEREBY REQUESTED BY THE UNDERMENTIONED APPLICANT  
ON THE BASIS OF THE PRESENT APPLICATION FILED IN DUPLICATE

INKOMSTE  
REPUBLIC VAN SUID AFRIKA  
370

A & A REF: 139808 AL

PATENT APPLICATION NO.	
21 01	989245
71	FULL NAMES(S) OF APPLICANT(S)

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54 TITLE OF INVENTION

"CROSS-LINKING OF POLYMERIC MATERIALS"

Only the items marked with an "X" in the blocks below are applicable.

- ☒ THE APPLICANT CLAIMS PRIORITY AS SET OUT ON THE ACCOMPANYING FORM P.2. The earliest priority claimed is Country:ZA No: 97/9107 Date: 10 OCTOBER 1997
- ☐ THE APPLICATION IS FOR A PATENT OF ADDITION TO PATENT APPLICATION NO. |21|01|
- ☐ THIS APPLICATION IS A FRESH APPLICATION IN TERMS OF SECTION 37 AND BASED ON APPLICATION NO. |21|01|

THIS APPLICATION IS ACCOMPANIED BY:

- ☒ A single copy of a provisional or two copies of a complete specification of 15 pages.
- ☐ Formal Drawings of sheets.
- ☐ Extra copy of drawings.
- ☒ Publication particulars and abstract (Form P.8 in duplicate) (for complete only).
- ☐ A copy of Figure of the drawings (if any) for the abstract (for complete only).
- ☐ An assignment of invention. (TO FOLLOW)
- ☐ Certified priority document(s) (State quantity): .....
- ☐ Translation of the priority document(s).
- ☐ An assignment of priority rights.
- ☒ A copy of Form P.2 and the specification of RSA Patent Application Nos. |21|01| 97/9107
- ☒ A Form P.2 in duplicate.
- ☐ A declaration and power of attorney on Form P.3. (TO FOLLOW)
- ☐ Request for ante-dating on Form P.4.
- ☐ Request for classification on Form P.9.
- ☐ Request for delay of acceptance on Form P.4.
- ☒ A copy of Form P.1

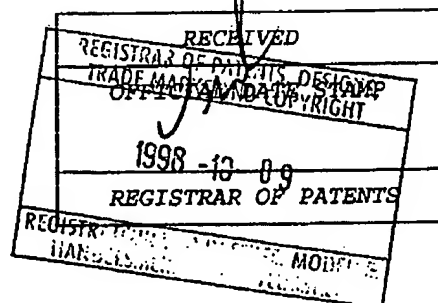
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DATED THIS 9th DAY OF OCTOBER 1998

A. LEWIS

ADAMS & ADAMS  
APPLICANTS PATENT ATTORNEYS

The duplicate will be returned to the applicant's address for service as proof of lodging but is not valid unless endorsed with official stamp.



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REPUBLIC OF SOUTH AFRICA  
Patents Act, 1978

## COMPLETE SPECIFICATION

(Section 30 (1) - Regulation 28)

OFFICIAL APPLICATION NO.

21 01

989245

LODGING DATE

22 9 October 1998

INTERNATIONAL CLASSIFICATION

51 C08J, B29D

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TITLE OF INVENTION

54 "CROSS-LINKING OF POLYMERIC MATERIALS"

**THIS INVENTION** relates to the cross-linking of polymeric materials. More particularly, this invention relates to a method of making partially cross-linked polymeric thermoplastic material suitable for use as a feedstock for the production of a film, to a partially cross-linked polymeric material suitable for use as a feedstock for the production of a film and to such film with specific shrink properties.

According to a first aspect of the invention there is provided a method of making a partially cross-linked polymeric thermoplastic material suitable for use as a feedstock for the production of a film, the method including the step of subjecting a polymeric thermoplastic starting material, which is capable of being cross-linked by ionising irradiation, to sufficient ionising radiation to cross-link the starting material partially.

The polymeric thermoplastic starting material may be any suitable polymeric thermoplastic material, for example a plastics, resinous or elastomeric [rubbery] material. The polymeric thermoplastic starting material may be selected from the group comprising homopolymers, co-polymers and ter-polymers of plastics materials, resinous materials, rubbers which are thermoplastic and blends thereof. Accordingly, the polymeric thermoplastic starting material may be selected from

the group consisting of:

polyethylene;

polyvinyl chloride;

segmented copolymers of butadiene and isoprene with styrene;

natural rubber;

blends of polyethylene, ethylene and propylene with rubber; and

mixtures of the foregoing.

The method may include the preliminary step of compounding or formulating the starting material as a mixture, by admixing with the starting material a suitable cross-linking agent. The starting material which is mixed with the cross-linking agent may be in the form of a continuous phase of the polymeric thermoplastic material and in which the cross-linking agent may be a discontinuous phase dispersed in finely divided form.

Usually the cross-linking agent will include at least two chemically reactive unsaturated bonds such as double bonds in its molecule, some of which unsaturated bonds react with the polymeric thermoplastic material during the partial cross-linking under the influence of the radiation. Suitable cross-linking agents include multifunctional acrylate or methacrylates such as trimethylolpropane trimethacrylate [TRIM] and allyl derivatives such as triallyl cyanurate [TAC].

The mixture may be compounded to contain 0.5-25% by mass of cross-linking agent. Compounding the mixture may take place in the usual way,

eg by mixing the constituents to a sufficient degree of homogeneity in the molten state at a temperature of typically not greater than 200°C and usually somewhat lower, and allowing the mixture to cool and solidify. Accordingly, in a particular embodiment, the method may include the preliminary step, before the irradiation, of compounding or formulating the starting material as a mixture by admixing a cross-linking agent into a molten polymeric thermoplastic material, the cross-linking agent including at least two unsaturated bonds in its molecule, which bonds are capable of reacting with the polymeric thermoplastic material under the influence of the irradiation to cross-link the polymeric thermoplastic material, the mixture being allowed to set before the irradiation is carried out.

The partial cross-linking may be effected by subjecting the starting material to ionising radiation, at ambient temperature. Alpha-, beta- or gamma radiation may be employed, and can be produced by a suitable radioactive radionuclide such as cobalt 60 or a suitable electron beam accelerator which generates energetic electrons with an energy of eg 250 keV to 10 MeV.

The absorbed radiation dose applied to said starting material during the partial cross-linking may be of the order of 0.1-100 kGy, and conventional electron beam machines or irradiators such as gamma irradiators can be employed for this purpose. In particular, the irradiation to which the starting material is subjected may be carried out at ambient temperature with the starting material in solid form by subjecting the starting material to sufficient radiation to apply an absorbed radiation dose to the starting material of 0.1-100 kGy.

The degree of cross-linking effected during the partial cross-linking by irradiation will depend on the nature of the starting material; and the cross-linking effected by the irradiation may be sufficient to achieve a degree of cross-linking ranging from 1-50% by mass of the material, as determined by Soxhlet solvent extraction. The degree of cross-linking effected may be such that further cross-linking of the material in further process steps is inhibited. It is to be understood that the degree of cross-linking effected is preferably such that heating of the feedstock in a film-blowing or casting step will not induce further cross-linking.

The starting material may be in powdered or pellet form during treatment with the ionising radiation.

The method may further include the step of, in addition to mixing the polymeric thermoplastic starting material with the cross-linking agent before the irradiation, admixing at least one additive therewith, also prior to the irradiation. The additive may be selected from the group which includes:

- plasticisers such as phthalic esters or epoxidised soya oil;
- impact modifiers such as ethylene-vinyl acetate copolymer or nitrile rubber;
- slip agents such as oleamide;
- fillers such as calcium carbonate;
- reinforcers such as fibres;
- flame retardants such as antimony trioxide and organobromine;
- coupling agents such as silanes;
- fungicides such as phosphites;

anti-blocking agents such as diatomaceous earth;  
 pigments such as rutile or  $TiO_2$ ;  
 essences such as dimethyl phenyl carbonyl;  
 lubricants such as stearic acid or calcium stearate;  
 blowing agents such as azodicarbonamide;  
 electronic conductors such as carbon black;  
 thermal stabilisers; and/or  
 UV stabilisers.

The additives used and the proportions thereof used will be similar to those conventionally employed for similar purposes in other polymeric materials.

Once the partial cross-linking has been effected, the partially cross-linked polymeric thermoplastic material can be used as a feedstock, optionally after certain additional method steps have been carried out thereon.

Thus, the method may include the step of admixing the partially cross-linked material with at least one stabiliser. The stabiliser may be in the form of an antioxidant such as hindered phenol octadecyl 3-(3,5-di-t-butyl-4-hydroxyphenyl) propionate, or the like. The stabiliser may be in the form of an ultra-violet radiation stabiliser such as poly-([6-((1,1,3,3-tetraethyl butyl)-imino)-1,3,5-triazine-2,4-diyl][2-(2,2,6,6-tetraethyl piperidyl)-imino])-hexamethylene-[4-(2,2,6,6-tetraethyl piperidyl)-imino]), or the like.



The feedstock may be used to produce film in the form of lay-flat film by using known blown film or cast film extrusion techniques.

In a preferred embodiment of the invention, the feedstock is used to produce a shrink-film suitable for packaging applications, such as collation packaging.

In another preferred embodiment of the invention, the feedstock is used to produce sheets suitable for use as shrink covering or coating for various products and material types where protection against corrosion and abrasion is required.

According to a second aspect of the invention there is provided a partially cross-linked polymeric thermoplastic material whenever made in accordance with the method as described above.

According to a third aspect of the invention, there is provided a film which includes a partially cross-linked polymeric thermoplastic material made in accordance with the method as described above.

The invention will now be described, by way of a non-limiting illustrative example, with reference to the following working example.

**EXAMPLE:**

A starting material in the form of a linear low density polyethylene (LLDPE) of grade LF 3101P, obtained in South Africa from Polyfin Limited, is irradiated to an absorbed dose of 6 kGy (not exceeding 10 kGy) in an industrial gamma irradiator to effect partial cross-linking.

The irradiated material is tumble-blended, at a final level of 2500ppm, with an anti-oxidant master batch which includes an anti-oxidant such Ultrinox 641, or a combination of primary, secondary and tertiary anti-oxidants. The stabiliser serves to protect the polymer both during and after extrusion into a film, thus enhancing the mechanical properties such as tear and impact strength of the film.

The tumble-blended material is extruded through a circular die to form an inflated bubble of film which is drawn away from the die to create a required degree of biaxial molecular orientation. Alternatively, the tumble-blended material is extruded through a slit die and then post extrusion orientated to achieve biaxial molecular orientation. The orientation or stress is frozen into the film during cooling used to solidify the molten material. In blown film the degree of orientation is controlled by balancing melt temperature, output rate from the circular die, draw-off rate from the circular die, size of bubble inflation and cooling rate. It is the frozen-in orientation which creates the film's shrink characteristics.

In cast film the degree of stretch applied to the film in machine and transverse directions defines the shrink characteristics.

The process as described may be used to produce a shrink-film having a thickness in the region of  $5\mu$  to  $5000\mu$ . In this particular example, a lay-flat shrinkfilm having a thickness in the region of  $50\mu$  is formed when using a blow ratio (which is calculated by a formula of  $0.637 \times [\text{lay-flat width} / \text{die diameter}]$ ) of 2.1:1.

The film is then slit into individual sheets of 320mm in length, each of which is wound on to a reel. The sheets are used for collation wrapping of canned food into required pack sizes, such as six-packs. In use, an appropriate length of the film is loosely wrapped around a group of six cans and the wrapped cans are then subjected to heat from a heat source such as a shrink tunnel or a gun which relieves the stress in the film so that the film shrinks tightly around the cans being wrapped, thereby holding them together.

One of the advantages of the invention, as described in the Example, is that a film is produced which has enhanced mechanical properties such as improved tear and impact strength.

**CLAIMS:**

1. A method of making a partially cross-linked polymeric thermoplastic material suitable for use as a feedstock for the production of a film, the method including the step of subjecting a polymeric thermoplastic starting material, which is capable of being cross-linked by ionising irradiation, to sufficient ionising radiation to cross-link the starting material partially.
2. The method as claimed in Claim 1, in which the polymeric thermoplastic starting material is selected from the group comprising homopolymers, co-polymers and ter-polymers of plastics materials, resinous materials, rubbers which are thermoplastic and blends thereof.
3. The method as claimed in Claim 2, in which the polymeric thermoplastic starting material is selected from the group consisting of:  
polyethylene;  
polyvinyl chloride;  
segmented copolymers of butadiene and isoprene with styrene;  
natural rubber;  
blends of polyethylene, ethylene and propylene with rubber; and  
mixtures of the foregoing.

4. The method as claimed in any one of Claims 1 to 3 inclusive, which includes the preliminary step of compounding or formulating the starting material as a mixture, by admixing with the starting material a suitable cross-linking agent.
5. The method as claimed in Claim 4, in which the starting material which is mixed with the cross-linking agent is in the form of a continuous phase of the polymeric thermoplastic material and in which the cross-linking agent is a discontinuous phase dispersed in finely divided form.
6. The method as claimed in Claim 4 or Claim 5, in which the cross-linking agent includes at least two chemically reactive unsaturated bonds in its molecule, some of which unsaturated bonds react with the polymeric thermoplastic material during the partial cross-linking under the influence of the radiation.
7. The method as claimed in any one of Claims 4 to 6 inclusive, in which the cross-linking agent includes a multifunctional acrylate.
8. The method as claimed in Claim 7, in which the acrylate is selected from the group including trimethylolpropane trimethacrylate [TRIM] and allyl derivatives such as triallyl cyanurate [TAC].
9. The method as claimed in any one of Claims 4 to 8 inclusive, in which the mixture is compounded to contain 0.5-25% by mass of the cross-linking agent.

10. The method as claimed in any one of the preceding claims, in which the partial cross-linking is effected by subjecting the starting material to ionising radiation, at ambient temperature.
11. The method as claimed in any one of the preceding claims, in which the absorbed radiation dose applied to the starting material during the partial cross-linking is of the order of 0.1-100 kGy.
12. The method as claimed in any one of the preceding claims, in which the cross-linking effected by the irradiation is sufficient to achieve a degree of cross-linking ranging from 1-50% by mass of the material, as determined by Soxhlet solvent extraction.
13. The method as claimed in Claim 12, in which the degree of cross-linking effected is such that further cross-linking of the material in further process steps is inhibited.
14. The method as claimed in any one of Claims 4 to 13 inclusive, which includes the step of, in addition to mixing the polymeric thermoplastic starting material with the cross-linking agent before the irradiation, admixing at least one additive therewith, also prior to the irradiation.
15. The method as claimed in Claim 14, in which the additive is selected from the group which includes:

plasticisers such as phthalic esters or epoxidised soya oil;  
impact modifiers such as ethylene-vinyl acetate copolymer or nitrile rubber;  
slip agents such as oleamide;  
fillers such as calcium carbonate;  
reinforcers such as fibres;  
flame retardants such as antimony trioxide and organobromine;  
coupling agents such as silanes;  
fungicides such as phosphites;  
anti-blocking agents such as diatomaceous earth;  
pigments such as rutile or  $TiO_2$ ;  
essences such as dimethyl phenyl carbonyl;  
lubricants such as stearic acid or calcium stearate;  
blowing agents such as azodicarbonamide;  
electronic conductors such as carbon black;  
thermal stabilisers; and  
UV stabilisers.

16. The method as claimed in any one of the preceding claims, which includes the step of admixing the partially cross-linked material with at least one stabiliser.

17. The method as claimed in Claim 16, in which the stabiliser is in the form of an antioxidant.

18. The method as claimed in Claim 17, in which the antioxidant is hindered phenol octadecyl 3-(3,5-di-t-butyl-4-hydroxyphenyl) propionate.

19. The method as claimed in Claim 16, in which the stabiliser is in the form of an ultra-violet radiation stabiliser.

20. The method as claimed in Claim 19, in which the ultra-violet radiation stabiliser is poly-([6-[(1,1,3,3-tetraethyl butyl)-imino]-1,3,5-triazine-2,4-diyl][2-(2,2,6,6-tetraethyl piperidyl)-imino])-hexamethylene-[4-(2,2,6,6-tetraethyl piperidyl)-imino]).

21. The method as claimed in any one of the preceding claims, in which the feedstock is used to produce film in the form of lay-flat film or cast film.

22. The method as claimed in any one of Claims 1 to Claim 21, inclusive, in which the feedstock is used to produce a shrink-film.

23. The method as claimed in any one of Claims 1 to Claim 21, inclusive, in which the feedstock is used to produce sheets of film suitable for use as a shrink covering or coating for various products and material types where protection against corrosion and abrasion is required.

24. A partially cross-linked polymeric thermoplastic material whenever made in accordance with the method as claimed in any one of Claims 1 to 20.




25. A film which includes a partially cross-linked polymeric thermoplastic material made in accordance with the method as claimed in any one of Claims 1 to 23.

26. A method of making a partially cross-linked polymeric thermoplastic material suitable for use as a feedstock for the production of a film, substantially as herein described with reference to the accompanying example.

27. A partially cross-linked polymeric thermoplastic material, substantially as herein described with reference to the accompanying example.

28. A film which includes a partially cross-linked polymeric thermoplastic material, substantially as herein described with reference to the accompanying example.

DATED this 9th day of OCTOBER 1998



**A. LEWIS**  
**ADAMS & ADAMS**  
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